Formation and Stability of Binary Complexes of Divalent Ecotoxic Ions (Ni, Cu, Zn, Cd, Pb) with Biodegradable Aminopolycarboxylate Chelants (dl-2-(2-Carboxymethyl)Nitrilotriacetic Acid, GLDA, and 3-Hydroxy-2,2′-Iminodisuccinic Acid, HIDS) in Aqueous Solutions

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Formation and stability of binary complexes of divalent ecotoxic ions (Ni, Cu, Zn, Cd, Pb) with biodegradable aminopolycarboxylate chelants (DL-2-(2-carboxymethyl)nitrilotriacetic acid, GLDA, and 3-hydroxy-2,2´-iminodisuccinic acid, HIDS) in aqueous solutions

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Abstract

The protonation and complex formation equilibria of two biodegradable aminopolycarboxylate chelants (DL-2-(2-carboxymethyl)nitrilotriacetic acid (GLDA) and 3-hydroxy-2,2´-iminodisuccinic acid (HIDS)) with Ni^{2+}, Cu^{2+}, Zn^{2+}, Cd^{2+} and Pb^{2+} ions was investigated using the potentiometric method at a constant ionic strength of $I = 0.10$ mol·dm^{-3} (KCl) in aqueous solutions at 25 ± 0.1°C. The stability constants of the proton-chelant and metal-chelant species for each metal ion were determined, and the concentration distributions of various complex species in solution were evaluated for each ion. The stability constants ($\log_{10}K_{\mathrm{ML}}$) of the complexes containing Ni^{2+}, Cu^{2+}, Zn^{2+}, Cd^{2+} and Pb^{2+} ions followed an identical order of $\log_{10}K_{\mathrm{CuL}} > \log_{10}K_{\mathrm{NiL}} > \log_{10}K_{\mathrm{PbL}} > \log_{10}K_{\mathrm{ZnL}} > \log_{10}K_{\mathrm{CdL}}$ when using GLDA (13.03 >12.74 >11.60 >11.52 >10.31) as when using HIDS (12.63 >11.30 >10.21 > 9.76 >7.58). In each case, the constants obtained for metal-GLDA complexes were higher in magnitude than the corresponding constants for metal-HIDS complexes. The conditional stability constants ($\log_{10}K'_{\mathrm{ML}}$) of the metal-chelant complexes containing GLDA and HIDS were calculated in terms of pH, and compared with the stability constants for EDTA and other biodegradable chelants.

Keywords: stability constant; biodegradable aminopolycarboxylate chelant; GLDA; HIDS; ecotoxic ions.

1.0 Introduction

Aminopolycarboxylate chelants (APCs) have been and continue to be extensively used in a variety of industrial processes [1, 2], including the treatment of toxic metal-contaminated solid waste materials [3-5]. APCs are commonly employed to restrict metal ions from playing their normal chemical roles through the formation of stable and water-soluble metal complexes [6, 7]. Because ethylenediaminetetraacetic acid (EDTA) forms stable water-soluble chelant complexes with the majority of toxic metals [2], it has been utilized most often among the APCs. The environmental consequences of the release of APCs to the surroundings has become an issue of concern despite their excellent metal-binding capacities [8]. Remobilization of metal ions from soils and sediments into the aqueous phase may occur when APCs are released into aquatic environments [2]. Lethal exposures resulting from the presence of APCs are likely to persist for a longer period of time because of their poor photo-, chemo- and biodegradability [9-11]. In most cases, an increase in the threshold values of the toxic effects may be observed upon metal complexation [12, 13]. APCs raise the total nitrogen content and phosphate solubility in interstitial waters, and thereby contribute to eutrophication [14, 15]. Legislative regulations have become increasingly stringent about the environmental release of APCs [16, 17], resulting in a wide range of proposals for the treatment of APC-containing wastewater [18, 19]. Alternatively, the search for alternatives to classical APCs in the form of eco-friendly biodegradable variants has become a topic of interest for the treatment of solid waste materials [20-22] or application in the chelant-enhanced phytoextraction of toxic metals [23, 24]. Several biodegradable chelating agents, such as nitrilotriacetic acid (NTA), iminodisuccinic acid (IDSA), [S,S]-ethylenediaminedisuccinic acid (EDDS), methylglycine diacetic acid (MGDA) are considered potential alternatives to EDTA for the aforementioned operations, and the corresponding formation and stability data about their metal-chelant binary complexes are available [25]. The development of the new eco-friendly chelants and the study of their complexation behavior are critical for evaluating the usefulness of these chelants in specific treatment operations [26-29]. DL-2-(2-carboxymethyl)nitrilotriacetic acid (GLDA) and 3-hydroxy-2,2´-iminodisuccinic acid (HIDS) (Fig. 1) are two new commercially available APCs that are supposed to possess eco-friendly characteristics. Furthermore, improved biodegradability of GLDA [30] and HIDS [31] relative to EDTA has been proposed. The complexation properties of these chelants have not been reported in detail in the standard reference databases of critically selected stability constants of metal complexes. This fundamental information is necessary for assessing new biodegradable chelants for use in a variety of chelant-based industrial clean-up and environmental remediation processes. Therefore, we report on the complexation behavior of GLDA and HIDS and divalent ecotoxic ions (Ni, Cu, Zn, Cd, and Pb) in aqueous solutions, which will be useful for the design of eco-friendly waste management processes.

2.0 Experimental Section

2.1 Instrumentation

KEM AT-610 automatic titrator (Kyoto Electronics, Kyoto, Japan), equipped with a pH-combination electrode and a temperature probe, was used for potentiometric measurements. The electrode system was calibrated with standard buffer solutions (pH 4.0, 7.0 and 9.0 prepared from buffer powders (Horiba, Kyoto, Japan) at 25 ± 0.1°C before and after each series of pH measurements. A 100 cm^3 titration vessel, equipped with a magnetic stirrer and a water-jacket type thermostat with a TAITEC EL-8F Coolnit bath water circulator (Saitama, Japan), was used to stir and maintain a constant temperature during the titration. The vessel was sealed with a special cover containing inlets for the electrode, temperature probe, and dosing nozzle for the titrator, in addition to a nitrogen gas inlet and outlet. Nitrogen gas was used to eliminate the ingress of CO_2 and maintain an inert atmosphere.

The iCAP 6300 inductively coupled plasma optical emission spectrometer from Thermo Fisher Scientific (Waltham, MA) was used to determine the metal concentration. The GLDA and HIDS concentrations were validated using an automated TOSOH 8020 high-performance liquid chromatography system from Tosoh (Tokyo, Japan). The Arium® Pro water purification system from Sartorius Stedim Biotech GmbH (Göttingen, Germany) was used to produce the ultrapure water (resistivity > 18.2 MΩ·cm).

2.2 Materials

GLDA from AkzoNobel (Amsterdam, Netherlands) and HIDS from Nippon Shukubai (Tokyo, Japan) were used in this study (Fig. 1). Both products were aqueous solutions of sodium salts, GLDA 40 wt% and HIDS 51.5 wt%. The products are commercially available and were used in the experiments without any additional treatment.

All of the chemicals and solvents used were of analytical reagent grade. Carbonate-free potassium hydroxide (Kanto Chemical, Tokyo, Japan) was standardized potentiometrically with potassium hydrogen phthalate (Wako Pure Chemical, Osaka, Japan). A solution of hydrochloric acid (Kanto Chemical, Tokyo, Japan) was standardized prior to use. Potassium chloride from Wako Pure Chemical (Osaka, Japan; > 0.99 mass fraction purity) was used to adjust the ionic strength of the system. Cadmium(II) chloride, copper(II) chloride dihydrate, nickel(II) chloride hexahydrate from Kanto Chemical (Tokyo, Japan; > 0.99 mass fraction purity), and Titrisol® ampoules of lead and zinc from Merck KGaA (Darmstadt, Germany) were used to prepare stock solutions of metals. "CO_2-free" water, used to prepare the working solutions, was obtained by boiling and cooling ultrapure water under a stream of nitrogen.

2.3 Software for computation

The computer program GLEE [32] was used to obtain an estimate of the carbonate concentration of the base by analyzing the results of strong acid-strong base titrations. GLEE was also used to confirm the concentration of the base and the pKw value (pKw = 13.78 at 25

± 0.1°C, I = 0.1 mol·dm^{-3}). The titration conditions were simulated with the HySS2009 program [33] prior to performing the titrations experimentally. The potentiometric data were analyzed using the HYPERQUAD 2008 program [34] to calculate the protonation and metal-chelant stability constants. The HYPERQUAD program facilitates the visual interpretation of refinement, in addition to providing a best fit for the titration data.

2.4 Estimation of protonation constants and metal-chelant stability constants

Aqueous solutions (A–D) of 50 cm^3 (total volume) were titrated with 0.1 mol·dm^{-3} KOH at 25 ± 0.1°C. The ionic strength of the solutions was maintained constant at 0.1 mol·dm^{-3} by the addition of an appropriate amount of 1.0 mol·dm^{-3} KCl stock solution.

Solution A: HCl (1.0 × 10^{-2} mol·dm^{-3}) + GLDA (1.0 × 10^{-3} mol·dm^{-3})

Solution B: HCl (1.0 × 10^{-2} mol·dm^{-3}) + GLDA (1.0 × 10^{-3} mol·dm^{-3}) + M(II) ions (M = Ni, Cu, Zn, Cd, Pb) (1.0 × 10^{-3} mol·dm^{-3})

Solution C: HCl (1.0 × 10^{-2} mol·dm^{-3}) + HIDS (1.0 × 10^{-3} mol·dm^{-3})

Solution D: HCl (1.0 × 10^{-2} mol·dm^{-3}) + HIDS (1.0 × 10^{-3} mol·dm^{-3}) + M(II) ions (M = Ni, Cu, Zn, Cd, Pb) (1.0 × 10^{-3} mol·dm^{-3})

Each solution was allowed to equilibrate for at least 30 minutes at 25 ± 0.1°C prior to performing the titration. The auto-titrator recorded the data at a constant volume increment and at pre-set intervals, producing a real-time titration curve. Each titration was repeated at least for three times, and more than 100 points of potentiometric measurements were utilized in the data analysis.

3.0 Results and Discussion

3.1 Protonation constants

The protonation constants for GLDA and HIDS were computed from the potentiometric pH profiles of the GLDA- and HIDS-spiked solutions in the absence of metal ions. Raw data for each titration were treated with a non-linear least-squares refinement using the

HYPERQUAD program, wherein the weights of the titrant are the independent variables and the pH values are the dependent variables. The percentage distribution of different protonation stages of GLDA and HIDS in the aqueous medium (I = 0.1 mol·dm^{-3}) at 25 ± 0.1°C is provided in Fig. 2. The proton-chelant constants for the overall reaction, β_n, can be described by the following relationship:

$$\beta_n = K_{a1} \cdot K_{a2} \cdots K_n = \frac{[H_nL]}{[H]^n[L]} \tag{1}$$

where K_{a1}, K_{a2}….K_n define the stepwise acid dissociation constants.

The overall (($\log_{10}\beta_{pqr}$) and successive ($\log_{10}K$) protonation constants for GLDA and HIDS, as calculated by the HYPERQUAD program, are provided in Tables 1 and 5, respectively. The species distribution curves of GLDA and HIDS (Fig. 2) demonstrate that the first protonation of L^{4-} to HL^{3-} occurs at the amino nitrogen atoms in an alkaline solution, and the HL^{3-} remains as the dominant species at pH 5.5–8.5 for HIDS (90–99.5%) and pH 6.0–8.4 (90–98.5%) for GLDA. The next protonations for GLDA (H_2L^{2-} to H_4L) and HIDS (H_2L^{2-} to H_5L^{+}) take place at the oxygen atoms of the carboxylate groups in the range of neutral to acidic pH. In GLDA, the association of the last proton occurs at the pH of 2, which is the lower limit of the pH range studied and therefore was not considered in the calculation. The predicted schemes of the protonation equilibria for GLDA and HIDS are provided in Figs. 3 and 4, and are found to be comparable with those reported for other chelants that have analogous structures [35-39]. The formation equilibria and protonation schemes of GLDA and HIDS demonstrate that the respective equilibrium constants depend on any or both of the following factors: (a) the effect of the substituent groups, (b) the space between the functional groups in the chelant structures.

The experimental protonation constant data for GLDA are fairly consistent with the data reported for the critically selected stability constants of metal complexes (shown in the parentheses of the Table 1) in the NIST database [25], despite the variation in the experimental conditions, such as ionic strength, background medium and methods of calculation. There are no data for HIDS in the NIST database.

3.2 Metal-chelant stability constants

The overall formation constants ($\log_{10}\beta_{pqr}$) for the binary systems containing metal ions (Ni^{2+}, Cu^{2+}, Zn^{2+}, Cd^{2+} and Pb^{2+}) and a chelant (GLDA or HIDS) at a molar ratio of metal ion (M) to chelant (L) of one to one were computed from the potentiometric titration data (Tables 2 and 3). The hydrolytic behavior (Table 4) of each metal species was taken into account when calculating the metal-chelant stability constants. The overall reaction can be represented by the following general equation:

$$p\mathrm{M} + q\mathrm{H} + r\mathrm{L} \Leftrightarrow \mathrm{M}_p\mathrm{H}_q\mathrm{L}_r \qquad \beta_{pqr} = \frac{[\mathrm{M}_p\mathrm{H}_q\mathrm{L}_r]}{[M]^p[H]^q[L]^r} \tag{2}$$

where p, q and r are the coefficients for metal ions, protons and chelants, respectively, which indicate the stoichiometry associated with the possible equilibria in solution.

The stepwise formation constant ($\log_{10}K$) for each of the species can be obtained from the differences between the various $\log_{10}\beta$ values. The $\log_{10}K$ values of GLDA and HIDS are provided in Table 5 and compared with those of NTA, IDSA, EDDS and EDTA. The stepwise formation equilibria can be defined by the following equations:

$$\mathrm{M} + \mathrm{L} \Leftrightarrow \mathrm{ML} \qquad K_{\mathrm{ML}} = \frac{[\mathrm{ML}]}{[\mathrm{M}][\mathrm{L}]} \tag{3}$$

$$\mathrm{ML} + \mathrm{H} \Leftrightarrow \mathrm{MHL} \qquad K^{\mathrm{H}}_{\mathrm{MHL}} = \frac{[\mathrm{MHL}]}{[\mathrm{ML}][\mathrm{H}]} \tag{4}$$

$$\mathrm{MHL} + \mathrm{H} \Leftrightarrow \mathrm{MH_2L} \qquad K^{\mathrm{H}}_{\mathrm{MH_2L}} = \frac{[\mathrm{MH_2L}]}{[\mathrm{MHL}][\mathrm{H}]} \tag{5}$$

Additional deprotonation reactions involving the coordination of water molecules can be defined by the following equation:

$$\mathrm{ML(H_2O)} \Leftrightarrow \mathrm{M(OH)L} + \mathrm{H} \qquad K_{\mathrm{M(OH)L}} = \frac{[\mathrm{M(OH)L}][\mathrm{H}]}{[\mathrm{ML(H_2O)}]} \tag{6}$$

The stoichiometries and stability constants of binary metal-chelant complexes were determined from a composition model that was consistent with the titration data, made sense from a chemical point of view, and offered a better statistical fit in comparison with other possible compositions. A good overlap was observed between the experimental and calculated pH values (graphical representations are available as the supplementary material), and the refinements of the data sets were obtained throughout the pH range for all the complexes.

Information about the actual metal-chelant species present in aqueous systems at different equilibrium conditions, which are controlled by the pH of the solution, and have a detrimental effect on the bioavailability of the metals and their corresponding physiological and toxicological behavior [40]. The formation of the protonated MH_2GLDA (M = Ni^{2+}, Zn^{2+}, Cd^{2+} and Pb^{2+}) and MH_2HIDS (M= Ni^{2+}, Cu^{2+}, Cd^{2+} and Pb^{2+}) at various pH values can be observed from the graphical distribution diagrams shown in Figs. 5a (I, III–V) and 5b (I, II, IV, V), respectively. $MHGLDA^{-}$, $MHHIDS^{-}$, $MGLDA^{2-}$ and $MHIDS^{2-}$ species were formed under acidic conditions in the presence of Ni^{2+}, Cu^{2+}, Zn^{2+}, Cd^{2+} and Pb^{2+}. The formation of stable mono-hydroxo complexes, $M(OH)GLDA^{3-}$ and $M(OH)HIDS^{3-}$, began under neutral conditions, except in the case of Ni^{2+} and GLDA (Fig. 5a-I). The formation of Cd_2HIDS was observed and is attributed to the lower coordination number of the metal ion than the number of the donor atoms in the HIDS chelant, or alternatively, as a result of steric hindrance [41]. The stability constant data obtained for the complexation between Cu(II) and GLDA are comparable to the data in the NIST database [25]. However, in the NIST database, there are no data for the GLDA complexation with Ni^{2+}, Zn^{2+}, Cd^{2+} and Pb^{2}. Furthermore, the data for HIDS are not included in the same database.

The stability of the metal-chelant complexes depends on a number of factors, including the oxidation state and coordination number of the metal ion, as well as the electronic structure and character of the chelant. These factors determine the nature of the bond between the metal and chelant, which may be an electrostatic or covalent interaction [42]. The stability of different ML complexes was in the order of $\log_{10}K_{CuL} > \log_{10}K_{NiL} > \log_{10}K_{PbL} > \log_{10}K_{ZnL} > \log_{10}K_{CdL}$ in the presence of both GLDA (13.03 > 12.74 > 11.60 > 11.52 > 10.31) and HIDS (12.63 > 11.30 > 10.21 > 9.76 > 7.58). The constants obtained for the metal-GLDA complexes, were found to be greater in magnitude than the corresponding constants for the metal-HIDS complexes.

The stability sequence for the Cu^{2+}, Ni^{2+} and Zn^{2+} complexes with GLDA or HIDS follows the Irving-Williams series [43]: Ni(II) < Cu(II) > Zn(II). The stability of the Pb^{2+} complex with GLDA or HIDS is higher than that of the Zn^{2+} and Cd^{2+} complexes. A similar trend was also observed for other chelants containing oxygen (of the carboxylic group) as the donor atom, such as TMS (1-hydroxy-3-oxapentane-1,2,4,5-tetracarboxylic acid) and TDS (3,6-dioxaoctane-1,2,4,5,7,8-hexacarboxylic acid) [44].

3.3 Conditional metal-chelant stability constants

The stepwise or overall formation constant provides fundamental information about the stability of a metal–chelant complex in solution [45]. However, these values do not include factors that are likely to affect the system, such as the pH or the presence of interferences from coexisting species, and are thus rarely applicable for practical purposes [46]. Therefore, the term ‘conditional stability constant’ is defined as the effect of side reactions that may occur during the complexation of chelant with metal ions, such as the effect of chelant protonation and hydrolysis that may occur when a metal ion is in solution [41]. Various expressions are available for defining the conditional stability constant ($\log_{10}K'_{ML}$), although the one most frequently used is the following [46]:

$$\log_{10} K'_{ML} = \log_{10} K_{ML} - \log_{10} \alpha_{HL} - \log_{10} \alpha_{M} \quad (7)$$

where $\log_{10}K_{ML}$ is the formation constant of the 1:1 metal–chelant species. Side reactions involving chelant protonation are expressed by the term α_{HL}. Other interfering reactions, as denoted by the term α_{M}, include the formation of metal hydroxides and the effect of buffers. The formation of metal-chelant-proton species (MLH) or the metal-chelant-hydroxide species (MLOH) may also influence the conditional constant for a particular pH and can be taken into account with the term α_{ML} in eq. (7):

$$\log_{10} K'_{\mathrm{ML}} = \log_{10} K_{\mathrm{ML}} - \log_{10} \alpha_{\mathrm{HL}} - \log_{10} \alpha_{\mathrm{M}} + \log_{10} \alpha_{\mathrm{ML}} \quad (8)$$

The form of the equation used for the calculation of conditional constant depends on the incorporation of necessary metal hydroxide species, metal-chelant-proton species or metal-chelant-hydroxide species in the computation at a set pH. Accordingly, eq. (7) is more frequently used than eq. (8) [46].

The $\log_{10}K'_{ML}$ values of the metal complexes with GLDA, HIDS and other chelants (NTA, IDSA, EDDS and EDTA) were calculated using the binary hydrolysis constants of the metal ions (Ni^{2+}, Cu^{2+}, Zn^{2+}, Cd^{2+} and Pb^{2+}) (Table 4) and the experimental or literature values of the equilibrium constants. The change in the $\log_{10}K'_{ML}$ values in terms of pH are illustrated in Fig. 6. The values of $\log_{10}K'_{ML} \geq 6$ are considered to be in the suitable complexation range for practical use and according to this scale, EDTA is appropriate for target metal ions in a wider pH range of 3 to 11. GLDA formed stable complexes of practical significance in the pH range of 4 to 11 with Cu^{2+} and Ni^{2+}, 5 to 11 with Pb^{2+}, and 6 to 11 with Cd^{2+} and Zn^{2+}. For HIDS, the pH range was 4–11 with Cu^{2+}, 5–11 with Ni^{2+}, 6–11 with Zn^{2+} and Pb^{2}, and 8–11 with Cd^{2+}. We observed that the stability of metal complexes with GLDA or HIDS is lower than that of EDTA, and these complexes also tend to form at a narrower pH range. However, the use of the biodegradable APCs is advantageous in terms of environmental safety. The relative stability of the metal-chelant complexes of GLDA, HIDS and the other biodegradable APCs (NTA, IDSA, EDDS) at the pH of 7 was EDDS > GLDA > NTA > HIDS > IDSA for Ni^{2+}, Cu^{2+}, Zn^{2+} and Pb^{2+}, and GLDA > EDDS > NTA > IDSA > HIDS for Cd^{2+}. The stability of metal complexes using HIDS was found to be lower than using GLDA, which indicates that the GLDA chelant is a better alternative to non-biodegradable APCs in comparison with HIDS. Furthermore, under neutral conditions, the complexation ability of GLDA is better than that of NTA and IDSA.

4.0 Conclusions

The complexion ability of two biodegradable APCs, namely GLDA and HIDS, with ecotoxic metal ions (Ni^{2+}, Cu^{2+}, Zn^{2+}, Cd^{2+} and Pb^{2+}) in aqueous solutions was investigated using experimental potentiometric analysis and simulated using the HYPERQUAD computer program. It was found that all the metal ions formed 1:1 complexes with GLDA and HIDS. The formation of mono- and di-protonated metal complexes occurred under acidic conditions, while mono-hydroxo complexes formed at a slightly alkaline pH. The conditional stability constants for GLDA and HIDS were calculated in the pH range of 2–11, and compared with those of EDTA and other biodegradable chelants (NTA, IDSA and EDDS). The metal-chelant complex stability for GLDA and HIDS was lower than that of EDTA, and exhibited a narrower working pH range. However, GLDA and HIDS have advantageous properties due to their lower post-operation ecotoxicity, and is the recommended choice compared with EDTA. The use of GLDA is also advised as the better biodegradable alternative relative to NTA and IDSA in a neutral environment.

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References

[1] Conway, M., Holoman, S., Jones, L., Leenhouts, R., Williamson, G.: Selecting and using chelating agents. Chem. Eng. **106**, 86–90 (1999)

[2] Nowack, B., VanBriesen, J.M.: Chelating agents in the environment In: B. Nowack and J. M. VanBriesen, (Eds.), Biogeochemistry of Chelating Agents, pp. 1–18. American Chemical Society, Washington, DC (2005)

[3] Raghavan, R., Coles, E., Dietz, D.: Cleaning excavated soil using extraction agents: A state-of-the-art review. J. Hazard. Mater. **26**, 81–87 (1991)

[4] Peters, R.W.: Chelant extraction of heavy metals from contaminated soils. J. Hazard. Mater. **66**, 151–210 (1999)

[5] Chang, F.-C., Lo, S.-L., Ko, C.-H.: Recovery of copper and chelating agents from sludge extracting solutions. Sep. Purif. Technol. **53**, 49–56 (2007)

[6] Leštan, D., Luo, C.L., Li, X.D.: The use of chelating agents in the remediation of metal-contaminated soils: a review. Environ. Pollut. **153**, 3–13 (2008)

[7] Hasegawa, H., Rahman, I.M.M., Kinoshita, S., Maki, T., Furusho, Y.: Non-destructive separation of metal ions from wastewater containing excess aminopolycarboxylate chelant in solution with an ion-selective immobilized macrocyclic material. Chemosphere **79**, 193–198 (2010)

[8] Rahman, I.M.M., Hossain, M.M., Begum, Z.A., Rahman, M.A., Hasegawa, H.: Eco-environmental consequences associated with chelant-assisted phytoremediation of metal-contaminated soil In: I. A. Golubev, (Ed.), Handbook of Phytoremediation. Nova Science Publishers, Inc., New York (2010)

[9] Egli, T.: Biodegradation of metal-complexing aminopolycarboxylic acids. J. Biosci. Bioeng. **92**, 89–97 (2001)

[10] Nowack, B.: Environmental chemistry of aminopolycarboxylate chelating agents. Environ. Sci. Technol. **36**, 4009–4016 (2002)

[11] Nörtemann, B.: Biodegradation of chelating agents: EDTA, DTPA, PDTA, NTA, and EDDS In: B. Nowack and J. M. VanBriesen, (Eds.), Biogeochemistry of Chelating Agents, pp. 150–170. American Chemical Society, Washington, DC (2005)

[12] Sillanpää, M., Oikari, A.: Assessing the impact of complexation by EDTA and DTPA on heavy metal toxicity using microtox bioassay. Chemosphere **32**, 1485–1497 (1996)

[13] Sorvari, J., Sillanpää, M.: Influence of metal complex formation on heavy metal and free EDTA and DTPA acute toxicity determined by *Daphnia magna*. Chemosphere **33**, 1119–1127 (1996)

[14] Horstmann, U., Gelpke, N.: Algal growth stimulation by chelatisation risks associated with complexants in P-free washing agents. Rev. Intl. Oceanogr. Med. **260**, 101–104 (1991)

[15] Hering, J.G., Morel, F.M.M.: Kinetics of trace metal complexation: role of alkaline-earth metals. Environ. Sci. Technol. **22**, 1469–1478 (2002)

[16] van Ginkel, C.G., Geerts, R.: Full-Scale biological treatment of industrial effluents containing EDTA In: B. Nowack and J. M. VanBriesen, (Eds.), Biogeochemistry of Chelating Agents, pp. 195–203. American Chemical Society, Washington, DC (2005)

[17] Grundler, O.J., van der Steen, A.T.M., Wilmot, J.: Overview of the European risk assessment on EDTA In: B. Nowack and J. M. VanBriesen, (Eds.), Biogeochemistry of Chelating Agents, pp. 336–347. American Chemical Society, Washington, DC (2005)

[18] Hasegawa, H., Rahman, I.M.M., Nakano, M., Begum, Z.A., Egawa, Y., Maki, T., Furusho, Y., Mizutani, S.: Recovery of toxic metal ions from washing effluent containing excess aminopolycarboxylate chelant in solution. Water Res. **45**, 4844–4854 (2011)

[19] Sillanpää, M.E.T., Agustiono Kurniawan, T., Lo, W.-h.: Degradation of chelating agents in aqueous solution using advanced oxidation process (AOP). Chemosphere **83**, 1443–1460 (2011)

[20] Tandy, S., Bossart, K., Mueller, R., Ritschel, J., Hauser, L., Schulin, R., Nowack, B.: Extraction of heavy metals from soils using biodegradable chelating agents. Environ. Sci. Technol. **38**, 937–944 (2004)

[21] Zhang, L., Zhu, Z., Zhang, R., Zheng, C., Zhang, H., Qiu, Y., Zhao, J.: Extraction of copper from sewage sludge using biodegradable chelant EDDS. J. Environ. Sci. **20**, 970–974 (2008)

[22] Tandy, S., Healey, J.R., Nason, M.A., Williamson, J.C., Jones, D.L.: Remediation of metal polluted mine soil with compost: Co-composting versus incorporation. Environ. Pollut. **157**, 690–697 (2009)

[23] Nowack, B., Schulin, R., Robinson, B.H.: Critical assessment of chelant-enhanced metal phytoextraction. Environ. Sci. Technol. **40**, 5225–5232 (2006)

[24] Quartacci, M.F., Irtelli, B., Baker, A.J.M., Navari-Izzo, F.: The use of NTA and EDDS for enhanced phytoextraction of metals from a multiply contaminated soil by *Brassica carinata*. Chemosphere **68**, 1920–1928 (2007)

[25] Martell, A.E., Smith, R.M., Motekaitis, R.J., Texas A&M University, College Station, TX (2004)

[26] Pihko, P.M., Rissa, T.K., Aksela, R.: Enantiospecific synthesis of isomers of AES, a new environmentally friendly chelating agent. Tetrahedron **60**, 10949–10954 (2004)

[27] Martins, J.o.G., Barros, M.T., Pinto, R.M., Soares, H.M.V.M.: Cadmium(II), lead(II), and zinc(II) ions coordination of *N*,*N*'-(S,S)bis[1-carboxy-2-(imidazol-4yl)ethyl]ethylenediamine: Equilibrium and structural studies. J. Chem. Eng. Data **56**, 398–405 (2011)

[28] Sari, H., Can, M., Macit, M.: Potentiometric and theoretical studies of stability constants of glyoxime derivatives and their nickel, copper, cobalt and zinc complexes. Acta Chim. Slov. **52**, 317–322 (2005)

[29] El-Sherif, A.A., Shoukry, M.M., van Eldik, R.: Complex-formation reactions and stability constants for mixed-ligand complexes of diaqua(2-picolylamine)palladium(II) with some bio-relevant ligands. Dalton T., 1425–1432 (2003)

[30] Dissolvine® GL Technichal Brochure, Akzo Nobel Amsterdam, The Netherlands (2004)

[31] Biodegradable Chelating Agent: HIDS, Nippon Shokubai, Osaka, Japan (2008)

[32] Gans, P., O'Sullivan, B.: GLEE, a new computer program for glass electrode calibration. Talanta **51**, 33–37 (2000)

[33] Alderighi, L., Gans, P., Ienco, A., Peters, D., Sabatini, A., Vacca, A.: Hyperquad simulation and speciation (HySS): A utility program for the investigation of equilibria involving soluble and partially soluble species. Coordin. Chem. Rev. **184**, 311–318 (1999)

[34] Gans, P., Sabatini, A., Vacca, A.: Investigation of equilibria in solution. Determination of equilibrium constants with the HYPERQUAD suite of programs. Talanta **43**, 1739–1753 (1996)

[35] Ichikawa, T., Sawada, K.: Protonation behavior and intramolecular interactions of α,ω-alkanediaminepolymethylenepolyphosphonates. B. Chem. Soc. Jpn. **70**, 829–835 (1997)

[36] Sanna, D., Bodi, I., Bouhsina, S., Micera, G., Kiss, T.: Oxovanadium(IV) complexes of phosphonic derivatives of iminodiacetic and nitrilotriacetic acids. J. Chem. Soc. Dalton, 3275–3282 (1999)

[37] Sawada, K., Duan, W., Ono, M., Satoh, K.: Stability and structure of nitrilo(acetate-methylphosphonate) complexes of the alkaline-earth and divalent transition metal ions in aqueous solution. J. Chem. Soc. Dalton, 919–924 (2000)

[38] Popov, K., Niskanen, E., Ronkkomaki, H., H. J. Lajunen, L.: ^{31}P NMR Study of organophosphonate protonation equilibrium at high pH. New J. Chem. **23**, 1209–1213 (1999)

[39] Buglyó, P., Kiss, T., Dyba, M., Jezowska-Bojczuk, M., Kozlowski, H., Bouhsina, S.: Complexes of aminophosphonates–10. Copper(II) complexes of phosphonic derivatives of iminodiacetate and nitrilotriacetate. Polyhedron **16**, 3447–3454 (1997)

[40] Angkawijaya, A.E., Fazary, A.E., Hernowo, E., Taha, M., Ju, Y.-H.: Iron(III), chromium(III), and copper(II) complexes of L-norvaline and ferulic acid. J. Chem. Eng. Data **56**, 532–540 (2011)

[41] Ringbom, A.: Complexation in Analytical Chemistry. Interscience Publishers, New York (1963)

[42] Bell, C.F.: Principles and Applications of Metal Chelation. Clarendon Press, Oxford (1977)

[43] Irving, H., Williams, R.J.P.: The stability of transition-metal complexes. J. Chem. Soc., 3192–3210 (1953)

[44] Motekaitis, R.J., Martell, A.E.: Potentiometry of mixtures: metal chelate stability constants of 1-hydroxy-3-oxapentane-1,2,4,5-tetracarboxylic acid and 3,6-dioxaoctane-1,2,4,5,7,8-hexacarboxylic acid. Inorg. Chem. **28**, 3499–3503 (1989)

[45] Martell, A.E., Hancock, R.D.: Metal Complexes in Aqueous Solutions. Plenum Press, New York (1996)

[46] Davidge, J., Thomas, C.P., Williams, D.R.: Conditional formation constants or chemical speciation data? Chem. Spec. Bioavailab. **13**, 129–134 (2001)

[47] Baes, C.F., Messmer, R.E.: The Hydrolysis of Cations. Wiley Interscience, New York (1976)

Table 1. The overall protonation constants ($\log_{10}\beta_{pqr}$) for GLDA and HIDS in the aqueous medium at the ionic strength, I = 0.1 mol·dm^{-3} and at 25 ± 0.1°C[a]

Protonation equlibria	p	q	r	$\log_{10}\beta_{pqr}$	SD
$GLDA^{4-} + H^+ \rightleftharpoons HGLDA^{3-}$	0	1	1	9.39 (9.36)	0.04
$HGLDA^{3-} + H^+ \rightleftharpoons H_2GLDA^{2-}$	0	2	1	14.40 (14.39)	0.03
$H_2GLDA^{2-} + H^+ \rightleftharpoons H_3GLDA^-$	0	3	1	17.89 (17.88)	0.03
$H_3GLDA^- + H^+ \rightleftharpoons H_4GLDA$	0	4	1	20.45 (20.44)	0.03
$HIDS^{4-} + H^+ \rightleftharpoons HHIDS^{3-}$	0	1	1	9.61	0.02
$HHIDS^{3-} + H^+ \rightleftharpoons H_2HIDS^{2-}$	0	2	1	13.68	0.02
$H_2HIDS^{2-} + H^+ \rightleftharpoons H_3HIDS^-$	0	3	1	16.76	0.02
$H_3HIDS^- + H^+ \rightleftharpoons H_4HIDS$	0	4	1	18.90	0.03
$H_4HIDS + H^+ \rightleftharpoons H_5HIDS^+$	0	5	1	20.50	0.04

[a] All the values were calculated from the potentiometric data using HYPERQUAD 2008 (n = 3). The symbols p, q and r are the coefficients indicating the stoichiometry associated with the possible equilibria in solution. The data in the parentheses are from the NIST database of critically selected stability constants of metal complexes [25].

Table 2. The overall formation constants ($\log_{10}\beta_{pqr}$) for M(II) + GLDA (M = Ni, Cu, Zn, Cd, Pb) in the aqueous medium at the ionic strength, I = 0.1 mol·dm^{-3} and at 25 ± 0.1°C[a]

Formation reactions	p	q	r	$\log_{10}\beta_{pqr}$	SD
Ni^{2+}					
$Ni^{2+} + GLDA^{4-} \rightleftharpoons NiGLDA^{2-}$	1	0	1	12.74	0.07
$Ni^{2+} + H^{+} + GLDA^{4-} \rightleftharpoons NiHGLDA^{-}$	1	1	1	17.12	0.06
$Ni^{2+} + 2H^{+} + GLDA^{4-} \rightleftharpoons NiH_2GLDA$	1	2	1	19.33	0.06
Cu^{2+}					
$Cu^{2+} + OH^{-} + GLDA^{4-} \rightleftharpoons Cu(OH)GLDA^{3-}$	1	–1	1	3.12	0.04
$Cu^{2+} + GLDA^{4-} \rightleftharpoons CuGLDA^{2-}$	1	0	1	13.03	0.04
$Cu^{2+} + H^{+} + GLDA^{4-} \rightleftharpoons CuHGLDA^{-}$	1	1	1	17.16	0.05
Zn^{2+}					
$Zn^{2+} + OH^{-} + GLDA^{4-} \rightleftharpoons Zn(OH)GLDA^{3-}$	1	–1	1	0.88	0.04
$Zn^{2+} + GLDA^{4-} \rightleftharpoons ZnGLDA^{2-}$	1	0	1	11.52	0.05
$Zn^{2+} + H^{+} + GLDA^{4-} \rightleftharpoons ZnHGLDA^{-}$	1	1	1	16.12	0.06
$Zn^{2+} + 2H^{+} + GLDA^{4-} \rightleftharpoons ZnH_2GLDA$	1	2	1	18.70	0.08
Cd^{2+}					
$Cd^{2+} + OH^{-} + GLDA^{4-} \rightleftharpoons Cd(OH)GLDA^{3-}$	1	–1	1	0.06	0.06
$Cd^{2+} + GLDA^{4-} \rightleftharpoons CdGLDA^{2-}$	1	0	1	10.31	0.05
$Cd^{2+} + H^{+} + GLDA^{4-} \rightleftharpoons CdHGLDA^{-}$	1	1	1	15.03	0.04
$Cd^{2+} + 2H^{+} + GLDA^{4-} \rightleftharpoons CdH_2GLDA$	1	2	1	18.49	0.04
Pb^{2+}					
$Pb^{2+} + OH^{-} + GLDA^{4-} \rightleftharpoons Pb(OH)GLDA^{3-}$	1	–1	1	0.95	0.08
$Pb^{2+} + GLDA^{4-} \rightleftharpoons PbGLDA^{2-}$	1	0	1	11.60	0.06
$Pb^{2+} + H^{+} + GLDA^{4-} \rightleftharpoons PbHGLDA^{-}$	1	1	1	16.29	0.08
$Pb^{2+} + 2H^{+} + GLDA^{4-} \rightleftharpoons PbH_2GLDA$	1	2	1	18.40	0.10

[a] All the values were calculated from the potentiometric data using HYPERQUAD 2008 (n = 3). The symbols p, q and r are the coefficients for metal ions, protons and chelants, respectively, indicating the stoichiometry associated with the possible equilibria in solution.

Table 3. The overall formation constants ($\log_{10}\beta_{pqr}$) for M(II) + HIDS (M = Ni, Cu, Zn, Cd, Pb) in the aqueous medium at the ionic strength, I = 0.1 mol·dm^{-3} and at 25 ± 0.1°C[a]

Formation reactions	p	q	r	$\log_{10}\beta_{pqr}$	SD
Ni^{2+}					
$Ni^{2+} + OH^{-} + HIDS^{4-} \rightleftharpoons Ni(OH)HIDS^{3-}$	1	–1	1	1.80	0.15
$Ni^{2+} + HIDS^{4-} \rightleftharpoons NiHIDS^{2-}$	1	0	1	11.30	0.14
$Ni^{2+} + H^{+} + HIDS^{4-} \rightleftharpoons NiHHIDS^{-}$	1	1	1	14.82	0.13
$Ni^{2+} + 2H^{+} + HIDS^{4-} \rightleftharpoons NiH_2HIDS$	1	2	1	17.06	0.14
Cu^{2+}					
$Cu^{2+} + OH^{-} + HIDS^{4-} \rightleftharpoons Cu(OH)HIDS^{3-}$	1	–1	1	3.68	0.15
$Cu^{2+} + HIDS^{4-} \rightleftharpoons CuHIDS^{2-}$	1	0	1	12.58	0.12
$Cu^{2+} + H^{+} + HIDS^{4-} \rightleftharpoons CuHHIDS^{-}$	1	1	1	16.23	0.11
$Cu^{2+} + 2H^{+} + HIDS^{4-} \rightleftharpoons CuH_2\ HIDS$	1	2	1	18.80	0.11
Zn^{2+}					
$Zn^{2+} + OH^{-} + HIDS^{4-} \rightleftharpoons Zn(OH)HIDS^{3-}$	1	–1	1	0.8	0.04
$Zn^{2+} + HIDS^{4-} \rightleftharpoons ZnHIDS^{2-}$	1	0	1	9.76	0.03
$Zn^{2+} + H^{+} + HIDS^{4-} \rightleftharpoons ZnHHIDS^{-}$	1	1	1	13.68	0.06
Cd^{2+}					
$Cd^{2+} + OH^{-} + HIDS^{4-} \rightleftharpoons Cd(OH)HIDS^{3-}$	1	–1	1	–2.62	0.09
$2Cd^{2+} + HIDS^{4-} \rightleftharpoons Cd_2HIDS$	2	0	1	10.22	0.29
$Cd^{2+} + HIDS^{4-} \rightleftharpoons CdHIDS^{2-}$	1	0	1	7.58	0.08
$Cd^{2+} + H^{+} + HIDS^{4-} \rightleftharpoons CdHHIDS^{-}$	1	1	1	12.69	0.17
$Cd^{2+} + 2H^{+} + HIDS^{4-} \rightleftharpoons CdH_2HIDS$	1	2	1	16.46	0.12
Pb^{2+}					
$Pb^{2+} + OH^{-} + HIDS^{4-} \rightleftharpoons Pb(OH)HIDS^{3-}$	1	–1	1	0.87	0.05
$Pb^{2+} + HIDS^{4-} \rightleftharpoons PbHIDS^{2-}$	1	0	1	10.21	0.05
$Pb^{2+} + H^{+} + HIDS^{4-} \rightleftharpoons PbHHIDS^{-}$	1	1	1	14.34	0.06
$Pb^{2+} + 2H^{+} + HIDS^{4-} \rightleftharpoons PbH_2HIDS$	1	2	1	16.75	0.08

[a] All the values were calculated from the potentiometric data using HYPERQUAD 2008 (n = 3). The symbols p, q and r are the coefficients for metal ions, protons and chelants, respectively, indicating the stoichiometry associated with the possible equilibria in solution.

Table 4. The overall formation constants ($\log_{10}\beta_{pq}$) for M(II) (M = Ni, Cu, Zn, Cd, Pb) complexes with OH^- at 25 ± 0.1°C [47][a]

Species M = Ni, Cu, Zn, Cd, Pb	p	q	$\log_{10}\beta_{pq}$ Ni^{2+}	Cu^{2+}	Zn^{2+}	Cd^{2+}	Pb^{2+}
$M(OH)^+$	1	–1	–10.06	–8.22	–9.15	–10.31	–7.86
$M(OH)_2$	1	–2	–19.22	–17.53	–17.10	–20.59	–17.27
$M(OH)_3^-$	1	–3	–13.01	–27.80	–28.39	–33.30	–27.99
$M(OH)_4^{2-}$	1	–4	–43.54	–39.12	–40.71	–46.91	–
$M_2(OH)^{3+}$	2	–1	–10.45	–	–8.89	–9.16	–6.16

[a] The symbols p and q are the coefficients for metal ions and protons, respectively, indicating the stoichiometry associated with the possible equilibria in solution.

Table 5. The protonation and complexation of the GLDA and HIDS with the metal ions (Ni^{2+}, Cu^{2+}, Zn^{2+}, Cd^{2+}, Pb^{2+}) compared with the corresponding values of NTA, IDSA, EDDS and EDTA in the aqueous medium at the ionic strength, I = 0.1 mol·dm^{-3} and at 25 ± 0.1°C

Equilibria	GLDA (H_4L)[a]	HIDS (H_4L)[a]	NTA (H_3L)[b]	IDSA (H_4L)[b]	EDDS (H_4L)[b]	EDTA (H_4L)[b]
	$\log_{10}K$	$\log_{10}K$	$\log_{10}K$	$\log_{10}K$	$\log_{10}K$	$\log_{10}K$
[HL]/[H][L]	9.36	9.61	9.46–9.84	10	10.01	9.52–10.37
[H_2L]/[HL][H]	5.01	4.07	2.52	4.24	6.84	6.13
[H_3L]/[H_2L][H]	3.49	3.08	(1.81)	3.24	3.86	2.69
[H_4L]/[H_3L][H]	2.56	2.14	(1.0)	1.97	2.95	2
[H_5L]/[H_4L][H]	–	1.6	–	–	–	(1.5)
[H_6L]/[H_5L][H]	–	–	–	–	–	(0.0)
Ni^{2+}						
[ML]/[MOHL][H]	–	9.5	10.86	–	–	(11.9)
[ML]/[M][L]	12.74	11.3	11.51	11.68	16.7	18.4
[MHL]/[ML][H]	4.38	3.52	–	4.14	3.22	3.1
[MH_2L]/[MHL][H]	2.19	2.24	–	–	–	(0.9)[c]
[ML_2]/[M][L]^2	–	–	16.32	–	–	–
Cu^{2+}						
[ML]/[MOHL][H]	9.91	8.9	9.2		10.38	(11.4)
[ML]/[M][L]	13.03	12.58	13	12.69	18.4	18.78
[MHL]/[ML][H]	4.13	3.65	1.6	4.01	3.48	3.1
[MH_2L]/[MHL][H]	–	2.57	–	2.65	1.95	2
[ML_2]/[M][L]^2	–	–	17.4	–	–	–
Zn^{2+}						
[ML]/[MOHL][H]	10.64	8.96	10.06	–	–	(11.6)
[ML]/[M][L]	11.52	9.76	10.65	9.88	13.4[e]	16.5
[MHL]/[ML][H]	4.6	3.92	–	4.29	6.68	3
[MH_2L]/[MHL][H]	2.58	–	–	–	2.48	(1.2)[c]
[ML_2]/[M][L]^2	–	–	14.27	–	–	–
Cd^{2+}						
[ML]/[MOHL][H]	10.25	10.2	11.25	–	–	(13.2)[c]
[ML]/[M][L]	10.31	7.58	9.76	8.33	10.9[e]	16.5
[MHL]/[ML][H]	4.72	5.11	–	4.68	4.5	2.9
[MH_2L]/[MHL][H]	3.46	3.77	–	3.28	–	(1.6)[c]
[ML_2]/[M][L]^2	–	–	14.47	–	–	–
[M_2L]/[ML][M]	–	2.64	–	–	–	–
Pb^{2+}						
[ML]/[MOHL][H]	10.65	9.34	–	–	–	–
[ML]/[M][L]	11.6	10.21	11.48	9.75	12.7[e]	18
[MHL]/[ML][H]	4.69	4.13	2.3[d]	–	5.9	2.8
[MH_2L]/[MHL][H]	2.11	2.41	–	–	–	(1.7)[c]
[MH_3L]/[MH_2L][H]	–	–	–	–	–	(1.2)[c]
[ML_2]/[M][L]^2	–	–	12.8[e]	16.27		–

[a] Calculated values from the experimental potentiometric data using HYPERQUAD 2008 (n = 3).
[b] From the NIST database of critically selected stability constants of metal complexes [25].
[c] I = 1 mol·dm^{-3}
[d] I = 0.5 mol·dm^{-3}
[e] At 20°C.

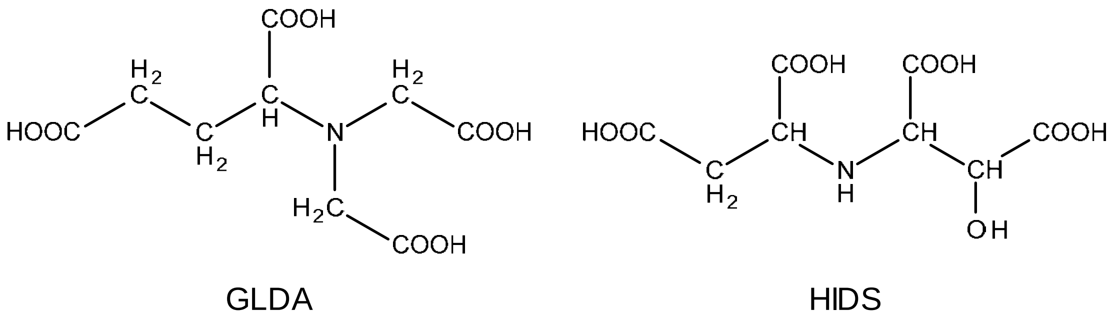

Figure 1. The chemical structures of DL-2-(2-carboxymethyl)nitrilotriacetic acid (GLDA) and 3-hydroxy-2,2´-iminodisuccinic acid (HIDS).

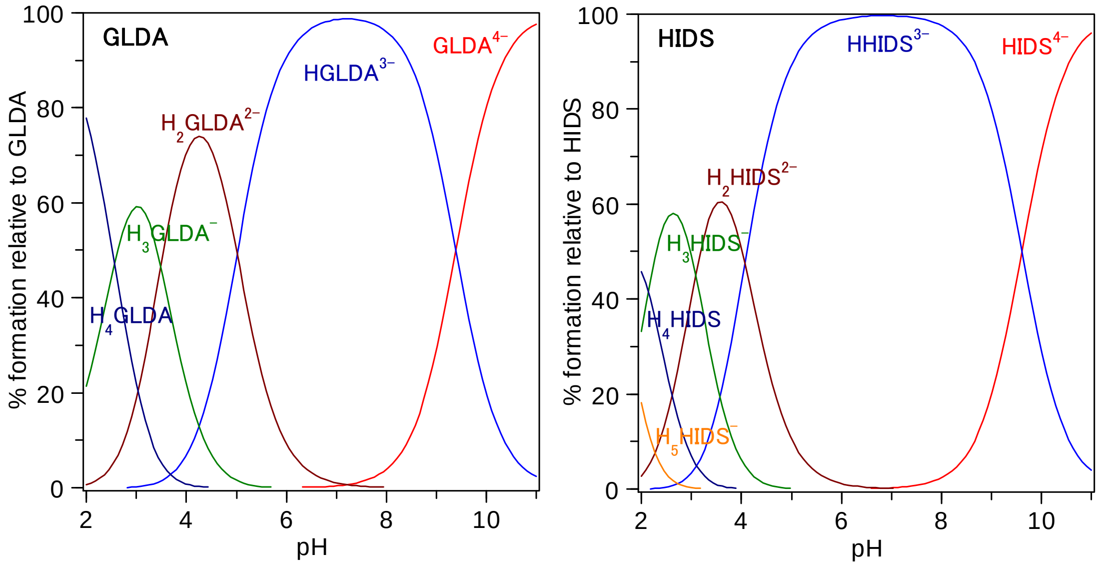

Figure 2. The percentage distribution of different protonation stages of GLDA and HIDS in the aqueous medium at the ionic strength, $I = 0.1$ mol·dm^{-3} and at 25 ± 0.1°C.

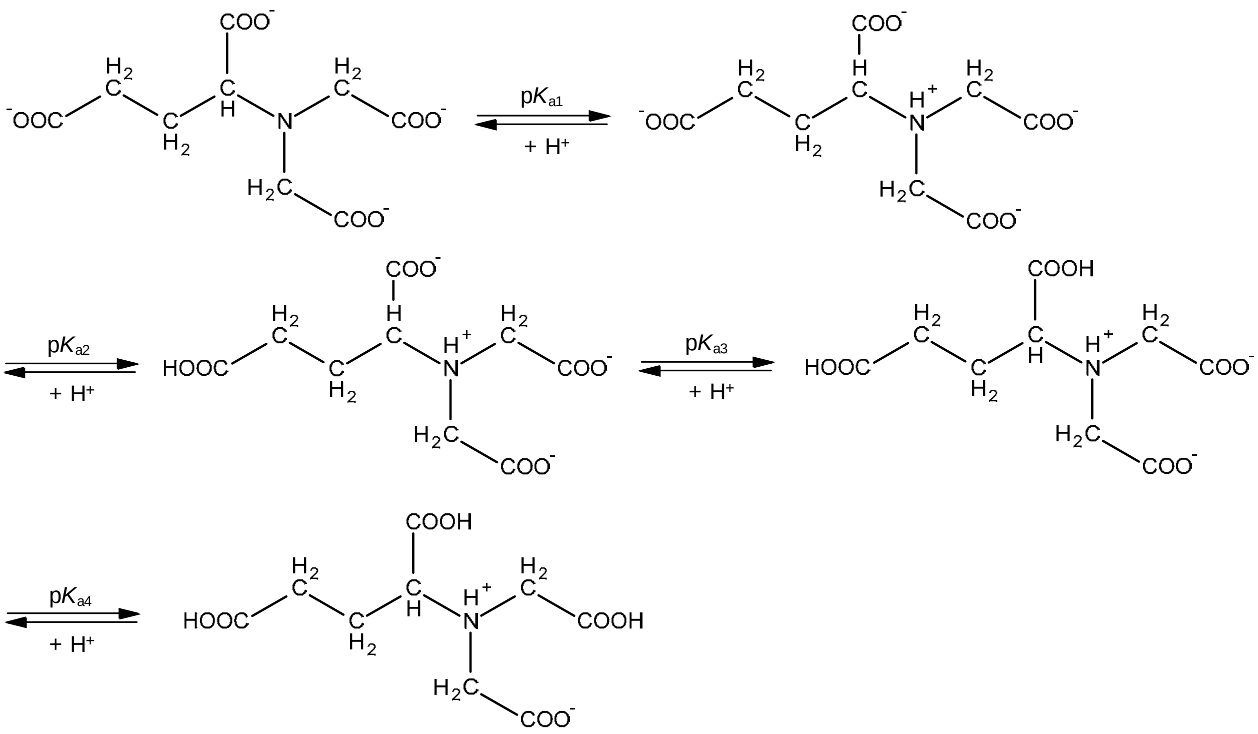

Figure 3. The predicted scheme of the protonation equilibria for GLDA in the aqueous medium at the ionic strength, I = 0.1 mol·dm^{-3} and at 25 ± 0.1°C.

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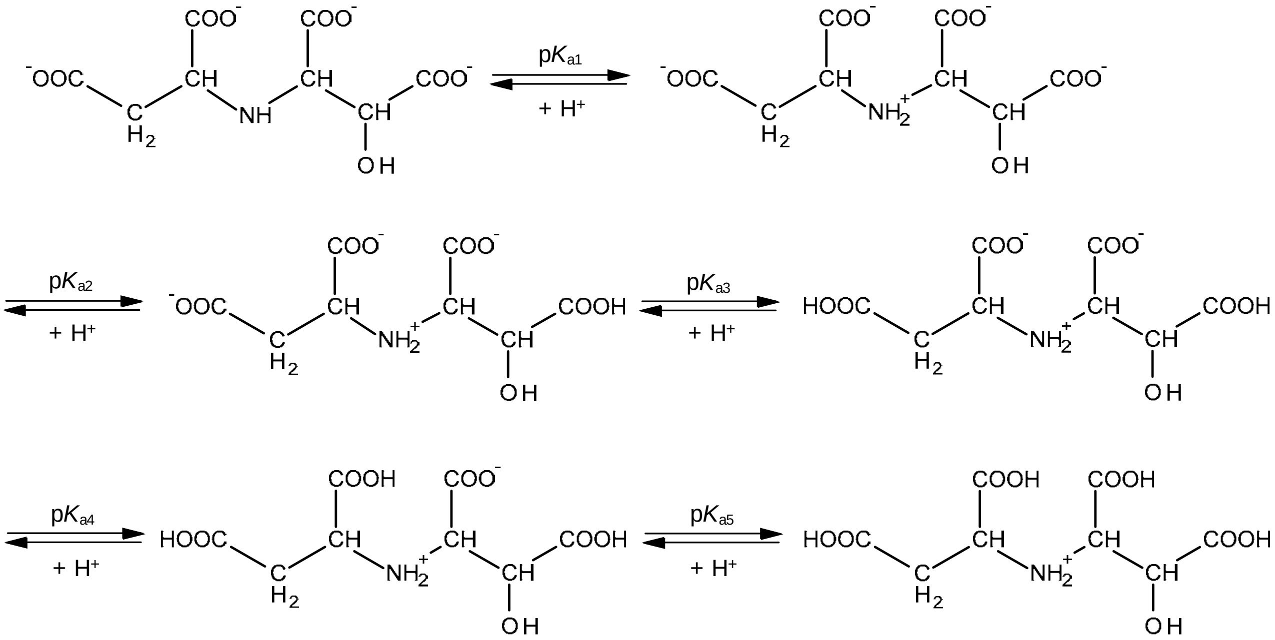

Figure 4. The predicted scheme of the protonation equilibria for HIDS in the aqueous medium at the ionic strength, $I = 0.1$ mol·dm^{-3} and at 25 ± 0.1°C.

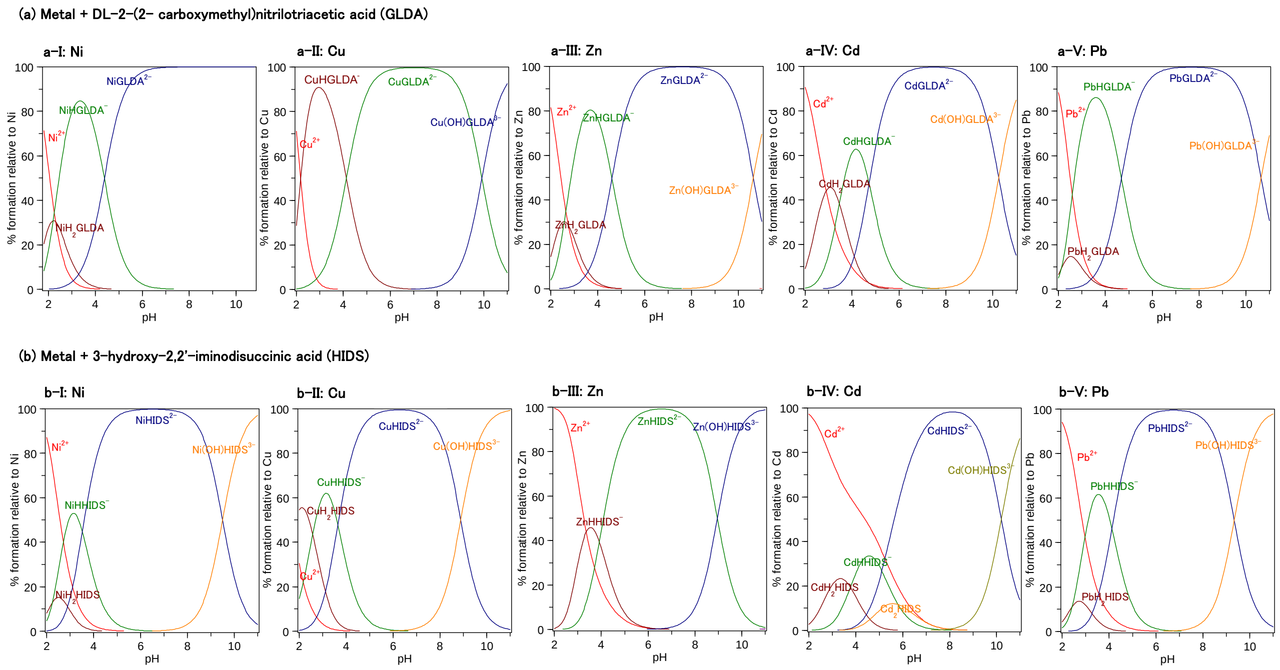

Figure 5. The species distribution curves for M(II) + L (M = Ni, Cu, Zn, Cd, Pb; L = GLDA or HIDS) in the aqueous medium at the ionic strength, $I = 0.1\ \text{mol}\cdot\text{dm}^{-3}$ and at 25 ± 0.1°C.

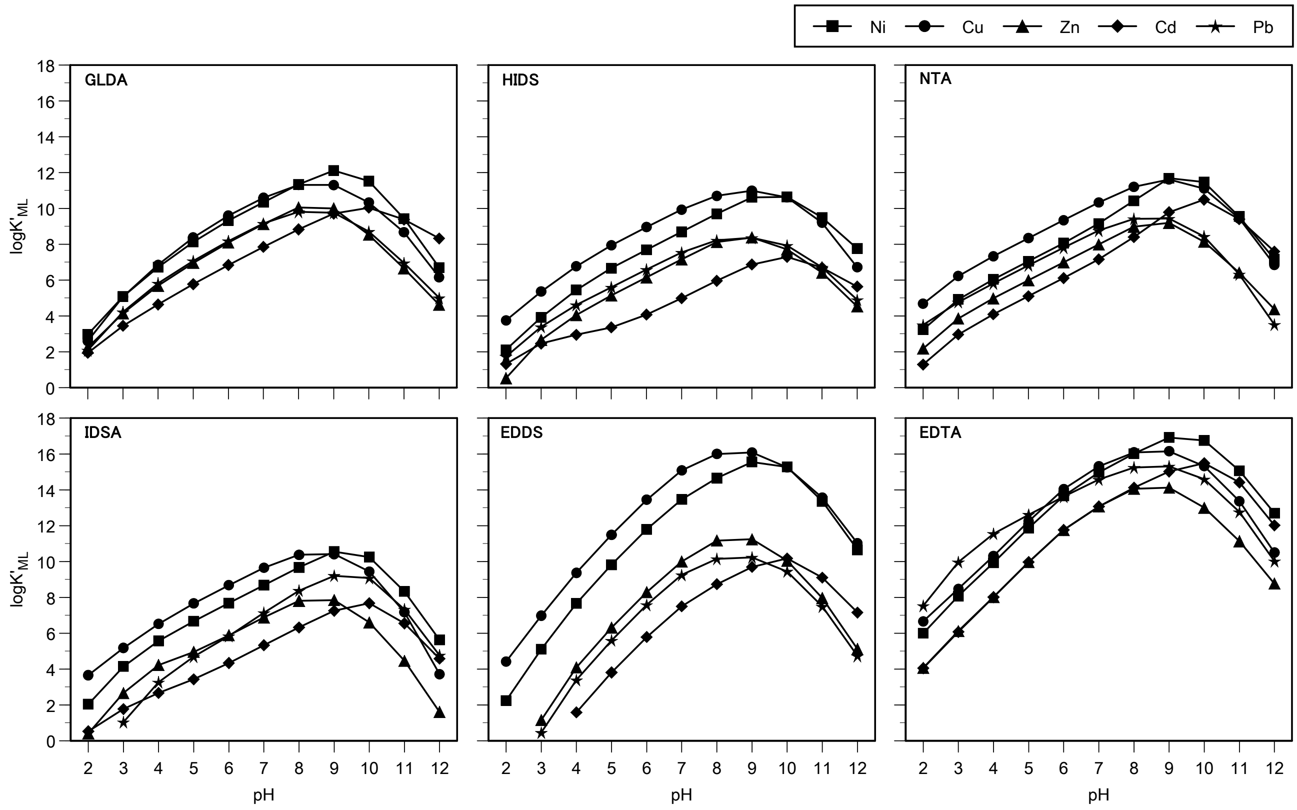

Figure 6. The conditional stability constants for M(II) + L (M = Ni, Cu, Zn, Cd, Pb; L = GLDA, HIDS, NTA, IDSA, EDDS, EDTA) in the aqueous medium as a function of pH.